On the Normal state electronic properties of Layered Sr_2RuO_4.

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Based on a comprehensive perusal of experimental results, we construct a microscopic model describing the normal state of layered $4d$ oxide Sr_2RuO_4 incorporating relevant quantum chemical features of the material. The high-T anomalies are explained within a Luttinger liquid (LL) picture. Interlayer one-particle hopping drives a dimensional crossover to a correlated Fermi liquid below a scale $E^* << t_\perp$. Using recently developed chain-dynamical mean field theory, the low value of E^*, as well as various puzzling features of the low-T normal state are explained as manifestations of the crossover from the high-T Luttinger liquid state in a consistent way.

Ruthenates constitute a class of $4d$ transition metal (TM) oxides existing in cubic as well as layered forms, and show a range of properties, from a ferromagnetic bad metal with a non-Fermi-liquid response (in $SrRuO_3$), via unconventional superconductivity (SC) in layered Sr_2RuO_4, to metamagnetic (quantum criticality?) in $Sr_3Ru_2O_7$[1] . These findings have led to intense activity to understand correlation effects in $4d$ TM oxides, in particular, the unconventional SC of Sr_2RuO_4[2].

Sr_2RuO_4 with Ru in a $4d^4$ state and a SC transition at $T_c \simeq 1.5K$[3], is isostructural to pure La_2CuO_4, however, *without* buckling of RuO_2 planes. The crystal field breaks the first Hund's rule, resulting in spin $S = 1$ at each Ru site. The tetragonal BCT structure seems to remain undistorted to the lowest T (the RuO_6 octahedra are elongated, leading to a lifting of the three-fold t_{2g} degeneracy). On structural and chemical grounds, one infers three electronically active bands: one (d_{xy}) two-dimensional band and two ($d_{yz,zx}$) 1D bands. These are indeed seen in band structure calculations[4], which give a Fermi surface seemingly in agreement with dHvA results[5]. Further, based upon early ideas [6], these calculations attempted to search for proximity to a ferromagnetic instability. Instead, proximity to *incommensurate* magnetic order (due to the almost nesting character of the 1D ($d_{yz,zx}$) bands was found, in nice agreement with recent inelastic neutron scattering (INS) results[7].

The agreement between LDA calculations and experiment stops as soon as finite energy/temperature responses are probed. At low-T (i.e $T_c < T < T^* = 30K$), the system shows all the characteristics of a correlated Fermi liquid (FL)[8]. The γ-coefficient of the low-T electronic specific heat yields a fourfold enhancement of the effective mass over the LDA prediction. Integrated photoemission spectra at low-T are quite different from the LDA results, but are well reproduced by inclusion of a dynamical correction (to second order) to the one-particle self-energy. The dc resistivity[8] shows anisotropic FL form, with a low residual value just above T_c. Careful perusal shows, however, that the ratio of "Woods-Saxon ratios" (along c and ab directions), $(A_c/\gamma^2) : (A_{ab}/\gamma^2) \simeq 1000$, a value too large to be related to a ratio of Fermi surface areas, indicating subtle influence of correlation effects also at lower T. Optical measurements[9] give more

evidence for anisotropic, correlated FL behavior at low-T.

Very interesting and unusual changes occur as T is raised above a characteristic scale $T^* \simeq 30K$. A smooth crossover to a non-FL metallic state (which shares many similarities with the non-FL normal state of underdoped HTC cuprates) is clearly revealed by experiment. The anisotropic dc resistivity shows the first sign of this change: $\rho_{ab}(T) \simeq A + BT$ for $T^* < T < 900K$, while $\rho_c(T) \simeq C_1 + C_2 T$ for $T^* < T < 120K$, with a crossover to an insulator-like form for $T > 120K$[9]. Clearly, the system falls into the "bad metal" class inspite of its low resistivity near T_c. Interesting facts are revealed by optical studies[9], where an extended "Drude" fit was used to analyze the low-energy response. Both the c-axis scattering rate and effective mass were found to have strong ω, T dependences. While $\tau_c^{-1}(\omega) \simeq a + b\omega$, the effective mass $m_c^*(\omega)$ has a near-logarithmic dependence for $T > T^*$. Below 30K, m_c^* increases to 40 times its band value at small energies, while $\tau_c^{-1}(\omega) \simeq a' + b'\omega^2$. However, the corresponding inplane $\tau_{ab}^{-1}(\omega)$ and $m_{ab}^*(\omega)$ are weakly ω-dependent, pointing to a direct correlation of the change of the electronic state with the change in c-axis dynamics.

Most interesting is the information obtained from recent INS studies[7], where a detailed analysis of the spin dynamics was carried out. At low-T, the magnetic fluctuations are found to be dominated by incommensurate peaks related to the Fermi surface nesting of the two quasi-1D bands. A pronounced softening in the spectrum near $\mathbf{Q_i} = (0.34, 0.34, 0)$ occurs upon cooling. Additionally, near $\mathbf{Q_i}$ and for $T > T^* = 30K$, $\chi''(\mathbf{q}, \omega)$ shows the so-called ω/T-scaling: $\chi''(\mathbf{q}, \omega) \simeq \omega^{-\gamma} f(\omega/T)$ with $\gamma = 0.75$ and $f(x) \propto x$ as $x \to 0$, reminiscent of similar behavior observed in certain rare-earth based heavy fermion compounds near their quantum critical points, but there essentially over the full Brillouin zone[10]. Hence the anomalous ω/T scaling found only near $\mathbf{q} = \mathbf{Q_i}$ in Sr_2RuO_4 must be ascribed to the proximity to the incommensurate AF instability involving the almost nested 1D bands. At low-T, however, a conventional Lorentzian form is adequate. Finally, the NMR relaxation rate shows a noticeable T dependence above 60K, with Korringa like behavior at lower T along with a sizable anisotropy ($\simeq 3$), implying an easy-axis spin anisotropy[7]. Thus

these results, taken together with[10] , reveal a *smooth* crossover from an anomalous high-T bad-metallic to a low-T anisotropic, renormalized FL state across T^*.

In this letter, we provide a unified understanding of these high-T anomalous features along with a description of the crossover to a FL using a material specific starting model. In particular, we show how specific quantum chemical features along with ubiquitous strong correlations are necessary ingredients for a unified understanding of the normal state. This is important because a detailed understanding of the nature of the normal state is crucial to pinpoint the "glue" that drives superconductive pairing.

Werner[11] has recently discussed the normal state physics of Sr_2RuO_4 from a similar starting point. Nevertheless, in what follows, we provide a very different theoretical modelling of the normal state that ties together all the above experimental results within a single approach.

To start, we notice that in Sr_2RuO_4, electron hopping involves Ru-4d-O-2p hybridization, which has a peculiar structure because of geometrical constraints imposed by lattice structure and t_{2g} orbital orientation. Considering an isolated RuO_2 plane to start with, it is easy to show[12] that the $4d_{xy} - 2p_{x,y}$ hybridization leads to a 2D band, and the $4d_{yz,zx} - 2p_{x,y}$ hybridization leads to two 1D bands, as found in LDA studies. In the undistorted, BCT structure, and without direct $O-O$ hopping, these three form mutually non-hybridizing bands. Small corrections arise from direct $O-O$ hopping: we do not consider them to begin with. This leads to an important conclusion: there is *no* interband mixing at one-particle level.

Local coulomb interactions, like the interorbital coulomb interaction $U_{\mu,\mu'}^{\sigma\sigma'}$ as well as the Hund's rule coupling J_H do lead to interband scattering processes. Finally, in Sr_2RuO_4, the spin-orbit $(s-o)$ coupling is important, as shown by the magnetic anisotropy[7]. This lifts the d-yz, zx orbital degeneracy and leads to a small mixing of the t_{2g} orbitals. So in the situation where the $s-o$ coupling (small interband one-particle hybridization) is irrelevant, the 1D correlations will dominate the physical response.

Strictly speaking, one should also consider the 2D-d_{xy} band in the analysis, but, since the singular effects of interactions are much stronger in 1D than in 2D (especially at fillings far away from commensurability), we do not consider the d_{xy} band at this point. However, it will become important once the dimensional crossover (see below) occurs at lower T.

Additional perturbations like interlayer one-particle hopping will lead to a dimensional crossover, restoring correlated FL behavior. We will consider these effects later below. The Hamiltonian describing an isolated RuO_2 plane is thus written as[11],

$$H = \sum_{<ij>,\mu,\mu',\sigma} t_{\mu\mu'}(c_{i\mu\sigma}^\dagger c_{j\mu'\sigma} + h.c) + H_{int} \qquad (1)$$

where

$$H_{int} = \sum_{i\mu\mu'\sigma\sigma'} U_{\mu\mu'}^{\sigma\sigma'} n_{i\mu\sigma} n_{i\mu'\sigma'} - \sum_{i,\mu,\mu'} \mathbf{S_{i\mu}} \cdot (J_H \mathbf{S_{i\mu'}} - \lambda \mathbf{L_{i\mu}}).$$

$$(2)$$

where μ, μ' represent the t_{2g} orbitals xy, yz, zx, with $U_{\mu\mu'}^{\sigma\sigma'} = U\delta_{\mu\mu'} + (1 - \delta_{\mu\mu'})[U_1\delta_{\sigma\sigma'} + U_2\delta_{\sigma,-\sigma'}]$ and the corresponding hopping parameters are taken from tight-binding fits to LDA results[4] , but $U, U_{\mu,\mu'}^{\sigma\sigma'}, J_H$ and λ must be regarded as parameters obtained by fitting to high-energy spectroscopic and magnetic data.

The bandstructure described above is modified in the presence of spin-orbit coupling. Indeed, the $s-o$ term couples states with different S_z. Making a unitary transformation to new operators which create (destroy) holes in the renormalized ground state containing contributions from the excited states of the original model via $s-o$ matrix elements, the hopping becomes spin-dependent, parametrized by: $t_{\mu\mu'}^{\sigma\sigma} = t, t_{\mu\mu'}^{-\sigma\sigma} = t\eta$, where η is treated as a parameter to fit the available magnetic data. The hopping part now reads,

$$H_0 = \sum_{<ij>,\mu,\mu',\sigma\sigma'} t_{\mu\mu'}^{\sigma\sigma'}(C_{i\mu\sigma}^\dagger C_{j\mu'\sigma'} + h.c) \qquad (3)$$

where $C_{i\sigma} = \sum_{\sigma'} V_{\sigma\sigma'} c_{i\sigma'}$, with $V_{\sigma\sigma'}$ a 2x2 unitary matrix. This rotation in spin space also produces new (four fermion) interaction terms in H_{int} above. However, these do not modify the special features of the hybridization, and so leave the low energy physics qualitatively unaffected. They should, however, be included in a strictly rigorous formulation.

From the above discussion, the dominant physical effects arise from the two 1D $(d_{zx,yx})$ bands, which are coupled to the 2D d_{xy} band by $U_{\mu,\mu'}^{\sigma\sigma'}, J_H$. From the structure of the hopping matrix, the d-xy, yz, xz electrons do not hybridize with each other but interact via $U_{\mu,\mu'}^{\sigma\sigma'}, J_H$, giving rise to additional strong scattering processes at low energy. This is an additional factor playing an important role in our description of the normal state.

Finally, to describe the 1D-3D crossover (which we will do later), we notice that the dominant coupling between the RuO_2 layers occurs via a one-particle tunnelling matrix element, $t_\perp$ between the 1D bands, since the d_{xy} orbitals have negligible overlap between neighboring layers. We mimic[11] the effect of finite J_H by setting $U_{\mu\mu'}^{\sigma,-\sigma} = U_1 > U_2 = U_{\mu\mu'}^{\sigma\sigma}$.

To bosonize the model represented by Eqs.(2-3), we notice that each of the (non-degenerate in presence of $s-o$ coupling) 1D $(d_{yz}$ for e.g.) bands with (equi-orbital) 1D hopping and U is modelled by a LL Hamiltonian with average band filling $n = 2/3$. This means that the 1D charge correlations can be modelled by a gaussian model for the charge bosonic field $\phi_\rho(x)$, since umklapp scattering is inoperative away from half-filling, and backscattering renormalizes to zero[13] for repulsive interactions. In addition, the inter-orbital interaction term $U_1 n_{id}^{yz} n_{id}^{zx}$ acts like a strong scattering potential for the yz band carriers

in the limit where the interband hybridization $t_{yz,zx}$ is effectively zero (a similar term comes from $U_1 n_{id}^{xy} n_{id}^{zx}$). There is an analogous effect coming from U_2. So in addition to the 1D LL physics in each of the yz, zx channels, one has to treat the infra-red singular effects (in 1D) arising from U_1 and U_2[14].

In the 1D case, spin-charge separation allows us to write down separate bosonized Hamiltonians for spin and charge collective modes. In our case relevant to Sr_2RuO_4, the final bosonized model for the zx band reads,

$$H_c^{(0)} = \int dx [K_\rho \Pi_\rho^2 + K_\rho^{-1}(\partial_x \phi_\rho(x))^2] \qquad (4)$$

with $H_{xre} = (U_1/(\pi a)^2) \int dx n_d^{yz} \partial_x \phi_\rho(x)$ from the interorbital interaction as described above. For the yz band, the same holds true with $x \to y$. These equations describing the 1D charge correlations are precisely those of "shifted" collective charge-density oscillations, and leave the LL behavior unchanged. They do, however, lead to an important rescaling of the Fermi-edge singularity (FES) exponent observed in knockout experiments with hard X-rays[14].

From the spin-dependent hopping above, one infers that the magnetic correlations are described by an effective anisotropic spin chain:

$$H_s = \sum_{<ij>} [J_z S_i^z S_j^z + (J_{xy} S_i^+ S_j^- + J_\perp S_i^+ S_j^+ + h.c)] \quad (5)$$

with $J_z = 4t^2(1+\eta)^2/U$, $J_{xy} = 4t^2(1-\eta^2)/U$ and $J_\perp = 4t^2\eta^2/U$ where the anisotropy parameter, $\Delta = (J_{xy}/J_z) = (1-\eta)/(1+\eta)$. The bosonized version of this model has the two-cosine structure of the XYZ model:

$$H_s = H_s^{(0)} - \int [\frac{m}{a\pi} cos(\beta_s \Phi_s) - \frac{m'}{a\pi} cos(\beta_s' \Theta_s)] dx. \quad (6)$$

where

$$H_s^{(0)} = \frac{u_s}{2} \int dx [(\Pi_s^2(x) + (\partial_x \phi_s)^2) \qquad (7)$$

This has a duality property[13]: when $J_z > |J_{xy}| - |J_\perp|$, J_{xy} scales down to zero, while $J_z, J_\perp \to \infty$. The dual field Θ_s gets ordered, leading to dominant SDW^{xy} correlations. When $J_z < |J_{xy}| - |J_\perp|$ (which for our case means $-2/3 < \eta < 0$) $J_\perp$ scales to irrelevance, and the resulting picture is qualitatively the same as for the XXZ model, and SDW^z correlations are dominant. In our case, we estimate $K_\sigma = 0.87$. In this regime, the SDW correlation function will be determined by both K_σ and the gapless *charge* sector, i.e., by K_ρ.

The spin correlation function is evaluated as a statistical average over phase variables[15], giving $\chi_s^{zz}(x) = < \mathbf{S}(x) \cdot \mathbf{S}(0) > \simeq \frac{cos(Q_i x)}{x^{K_\sigma^{-1}+K_\rho}}$ for the equal-time part. At finite T, and near $\mathbf{q} = \mathbf{Q}_i$, one obtains $\chi^{zz}(Q_i, T) \simeq T^{-K_\sigma^{-1}+K_\rho}$. The transverse dynamical spin susceptibility is given by

$$\chi_\perp(\mathbf{q}, \omega) \simeq \frac{A_\perp}{T^{K_\sigma^{-1} - K_\rho}} \frac{\omega}{T} \qquad (8)$$

and it goes like $\chi_\perp(\mathbf{q}, \omega) \simeq T^{-\gamma} \frac{\omega}{T}$ for $q' = (q - Q_i) \simeq 0$, with $\gamma = (K_\sigma^{-1} - K_\rho)$. The NMR relaxation rate, $1/T_1$ follows directly as

$$\frac{1}{T_1} = \frac{T}{\omega} \sum_{\mathbf{q}} \chi''(\mathbf{q}, \omega) \propto T^{-\gamma}. \qquad (9)$$

Finally, to compute the value of the charge stiffness K_ρ, we consider the Hamiltonian

$$H_c = H_c^{(0)} + \frac{U_1}{(a\pi)^2} \int (n_d^{xy} + n_d^{yz}) \partial_x \phi_\rho(x) dx. \qquad (10)$$

where $u_\rho K_\rho = v_F$ and $u_\rho / K_\rho = v_F + U$. In the intermediate coupling regime, $U \simeq 2.1$eV, $v_F = 0.7$eV[4], and $K_\sigma = 0.8$, one gets $K_\rho \simeq 1/2$ giving $\gamma = 0.65$ and the LL Fermi surface exponent, $\alpha = 1/8$. On the other hand, $K_\rho = 0.4$ (corresponding to $U \simeq 2.4$eV) yields $\gamma = 0.75$ and $\alpha = 0.23$. The value $\gamma = 0.75$ gives good fits to the quantum-critical scaling behavior of $\chi''(\mathbf{q} = \mathbf{Q}_i, \omega)$ above T^*.

The NMR relaxation rate is then $1/T_1 \propto T^{-0.75}$ which is not inconsistent with the observed T-dependence for $T \geq 100$K. Eqn.(7) with $\gamma = 0.75$ is also completely consistent with the experimental result, however, for $T > 40 - 50$K. At lower-T, $\chi(0, T)$ levels off and approaches a constant, consistent with a crossover to correlated FL behavior[16]. The in-plane dc resistivity shows almost linear behavior for $50K < T < 900K$, consistent with the Luttinger liquid physics above 50 K. More support for this picture comes from optics[17] where a crossover from incoherent response to a Drude-like response is indeed observed as T is lowered; however, interestingly, the scattering rate extracted from a generalized Drude fit shows T^2 dependence only up to $\simeq 30$K, in agreement with INS results.

To consider the LL-FL crossover as a function of T, we notice that the interlayer hopping, $t_\perp \simeq 0.02$eV (200K)[4]. A description of the effect of $t_\perp$ requires consideration of a model with coupled RuO_2 layers. Since the interlayer hopping for the d_{xy} band is much smaller than for the $d_{yz,zx}$ bands in the undistorted BCT structure, we are led to consider the model of coupled chains:

$$H = \sum_\nu H_{1D}^\nu - \sum_{i,\nu,\nu',\sigma} t_\perp (C_{i\nu\sigma}^\dagger C_{i\nu'\sigma} + h.c) \qquad (11)$$

for each of the 1D ($\nu = yz, zx$) bands. A description of the crossover by perturbation theory in $t_\perp$ is beset with difficulties, and is valid in the LL regime, but fails to reproduce the FL regime. Perturbation approaches in interaction work in the FL regime, but fail in the LL regime. An attractive way out is provided by a recent non-trivial extension of dynamical mean-field theory (DMFT) that replaces a single site by a single chain connected via $t_\perp$ to $z_\perp$ nearest neighbors, with

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$z_\perp \to \infty$[18]. Rigorously, one needs a numerical solution for the *full* single-chain propagator $G(\mathbf{k}, \omega)$, which is a very hard task, when considered together with the usual DMFT selfconsistency[18]. Fortunately, several conclusions can still be drawn without attempting a full solution. In the 1D regime, the in-chain self-energy, $\Sigma(k, \omega) \simeq t((k,\omega)/t)^{1/(1-\alpha)}$. From the DMFT equations, $t_\perp$ becomes relevant when $t_\perp > \Sigma$, yielding the crossover scale, $E^* \simeq t_\perp(t_\perp/t)^{\alpha/(1-\alpha)}$. In our case, this gives $E^* \simeq 60$K, qualitatively in agreement with, but somewhat higher than $T^* \simeq 30 - 40$K from experiment.

At $T < E^*$, chain-DMFT leads to anisotropic FL behavior. In particular, when $t_\perp << t$ and at low energies, all one-particle quantities obey the scaling $\omega' = \omega/E^*, k' = kE^*/t$ and $T' = T/E^*$; i.e., $t\Sigma(k, \omega, T) = E^*t_\perp\Sigma'(k', \omega', T')$ and $tG(k, \omega, T) = (E^*/t_\perp)G'(k', \omega', T')$ where Σ and G are universal functions associated with the crossover. A low-frequency expansion of Σ in the FL regime gives the quasiparticle residue $Z \simeq (t_\perp/t)^{\alpha/(1-\alpha)} = E^*/t_\perp$. Unlike in a conventional FL, this bears no resemblance to the effective mass enhancement, since both $(\partial\Sigma/\partial k)$ and $(\partial\Sigma/\partial\omega)$ scale in the same way. The interchain resistivity, $\rho_\perp(T)/\rho_0 = (t/E^*)R(T/E^*)$ with $R(x << 1) \propto x^2$ and $R(x >> 1) \propto x^{1-2\alpha}$. And the resistivity enhancement, $\rho_\perp(T)/\rho_0 = A(T/t)^2$ with $A = (t/t_\perp)^{3/(1-\alpha)}$. The resulting anisotropy of the Woods-Saxon ratio, $A_c/A_{ab} = (a/c)^2 A \simeq 1000$ for $\alpha = 0.23$, which is indeed in the right range[9]. Finally, the c-axis optical response is incoherent above E^*, with a coherent feature carrying a relative weight $\simeq Z^2$ appearing at low-T, again in qualitative agreement with observations[10]. An obvious inference from the above is that increasing T should lead to a disappearance of the quasicoherent features in photoemission. This may already have been observed experimentally[19].

At low-T, in the correlated FL regime, usual DMFT should provide a consistent description of electronic correlations. Such a program has been implemented[20] for the multiband system of the t_{2g} bands coupled by $U_{\mu\mu'}^{\sigma\sigma'}, J_H$. With $U = 2.5$eV, $J_H = 0.4$eV, the effective mass enhancement (from the self-energy) is $m^*/m \simeq 3 - 4$, completely consistent with specific heat data and dHvA measurements. In our picture, therefore, the effective mass enhancement arises from renormalization effects caused by local electronic correlations (DMFT) and has a very different origin from the one proposed by Werner[11]. Interestingly, this approach[20] also reconciles the apparent conflict between ARPES and dHvA data at low-T.

To conclude, starting from a material specific model for the layered TM oxide Sr_2RuO_4, we have described how the various high T anomalous features can be understood within Luttinger liquid ideas. The effects of $s - o$ coupling, necessary to obtain consistency with magnetic data, are consistently incorporated, leading to a new modelling for the high-T LL state compared to that of[11]. Finally, using the Luttinger liquid exponents obtained there, a *smooth* crossover to an anisotropic, 3D, correlated FL metallic state (below $E^* = 60$K) is obtained within the recently developed chain-DMFT. The low-T specific heat and static spin susceptibility are enhanced by conventional FL renormalization below T^*. Various thermodynamic and transport properties, some inexplicable within conventional scenarios, find a consistent explanation as manifestations of the interplay between the high-T LL state (irrelevance of $t_\perp$) and the low-T correlated FL state.

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